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Title: Isotopic Composition Measurements Using Ratio Based Method

Author(s): Vo, Duc Ta

Intended for: Training course

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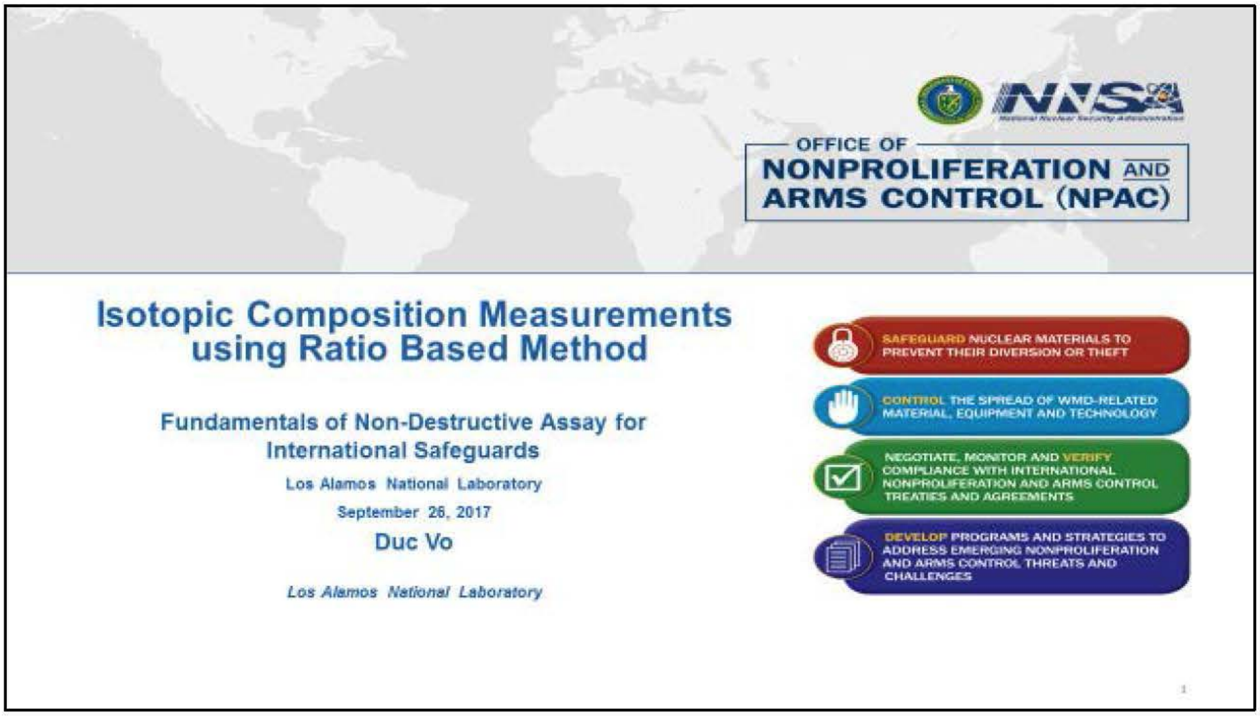

OFFICE OF
NONPROLIFERATION AND
ARMS CONTROL (NPAC)
Isotopic Composition Measurements using Ratio Based Method
Fundamentals of Non-Destructive Assay for International Safeguards
Los Alamos National Laboratory
September 26, 2017
Duc Vo
Los Alamos National Laboratory
SAFEGUARD NUCLEAR MATERIALS TO PREVENT THEIR DIVERSION OR THEFT
CONTROL THE SPREAD OF WMD-RELATED MATERIAL, EQUIPMENT AND TECHNOLOGY
NEGOTIATE, MONITOR AND VERIFY COMPLIANCE WITH INTERNATIONAL NONPROLIFERATION AND ARMS CONTROL TREATIES AND AGREEMENTS
DEVELOP PROGRAMS AND STRATEGIES TO ADDRESS EMERGING NONPROLIFERATION AND ARMS CONTROL THREATS AND CHALLENGES

Estimated Module Duration: 40 minutes

Required Tools and Materials:

1. Projector, screen, laptop with Word and PowerPoint programs
2. Participant guides, with slides and supplemental material

References:

1. Passive Nondestructive Assay of Nuclear Materials (PANDA): http://www.lanl.gov/orgs/n/n1/panda/
2. FRAM Manual (included in C:\FRAMData\References)

Supporting Documents:

1. None

Job Aids:

1. None

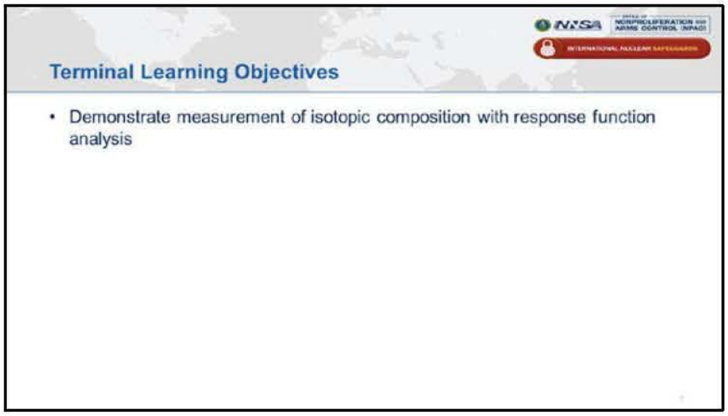
Terminal Learning Objectives (TLOs):

- TLO-1: Demonstrate measurement of isotopic composition with response function analysis

Enabling Learning Objectives (ELOs):

- ELO-1: State the categories of uranium and plutonium
- ELO-2: Discuss basic principles of response function analysis
- ELO-3: Describe the relative efficiency determination
- ELO-4: List conditions required for response function analysis
- ELO-5: Measure plutonium and uranium isotopic composition

Additional Information for Students:

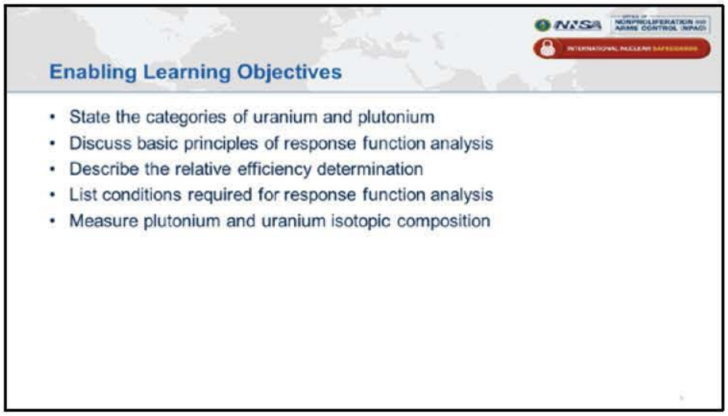


This lecture is focused on the isotopic ratio method. In the lab, we will use two different analysis programs: MGA/MGAU (developed by LLNL) and FRAM (developed by LANL).

Instructor Notes:

Review learning objectives with the participants.
Encourage participants to ask questions during the lecture.

Additional Information for Students:



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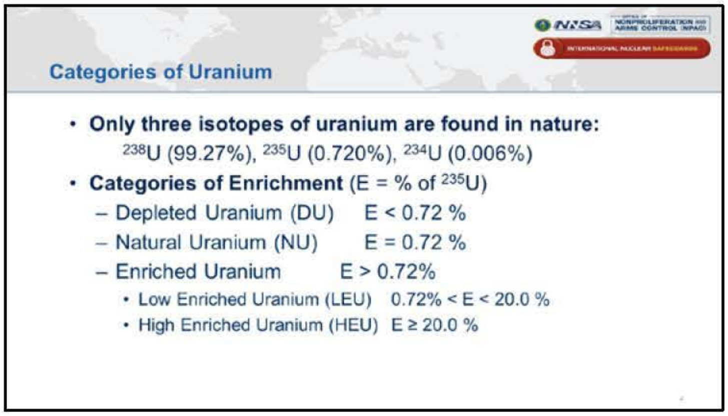
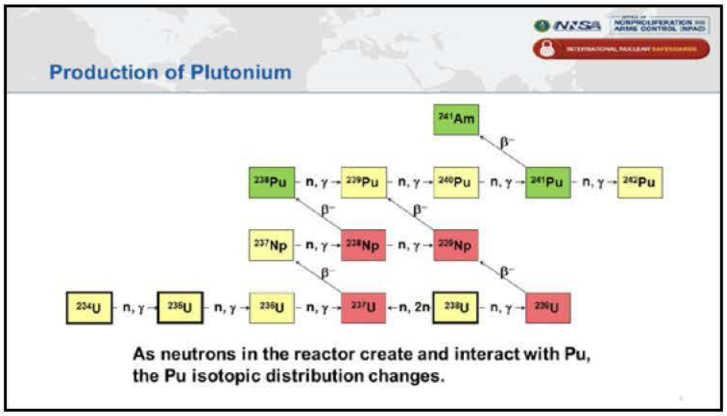


Instructor Notes:

Natural uranium constitutes of three naturally occurring isotopes: ^{238}U, ^{235}U and ^{234}U. Their Natural Abundance (NA), referred as the abundance of isotopes of a chemical element as naturally found on a planet, is respectively, 99.27%, 0.720% and 0.006%. ^{238}U is the most abundant and has the longest lifetime (4.5 billion years). It is not very radioactive and neutron capture by this nucleus leads to the formation of fissile ^{239}Pu in a reactor.
^{235}U is the only fissile nucleus found in natural uranium. ^{234}U is found in a very small percentage and occurs as an indirect decay product of ^{238}U.
Depleted uranium has a lower content of ^{235}U (<0.72%) than natural uranium. Most DU comes as a by-product of the production of enriched uranium for use as fuel in nuclear reactors and in the manufacture of nuclear weapons.
Enriched uranium (EU) is used in nuclear reactors that need a higher concentration (enrichment) of ^{235}U (>0.72%) than which exists in natural uranium. There are two types of enrichment: Low Enriched Uranium (LEU) (most commercial reactor fuel is enriched to 5%) and Highly Enriched Uranium (HEU).

Additional Information for Students:

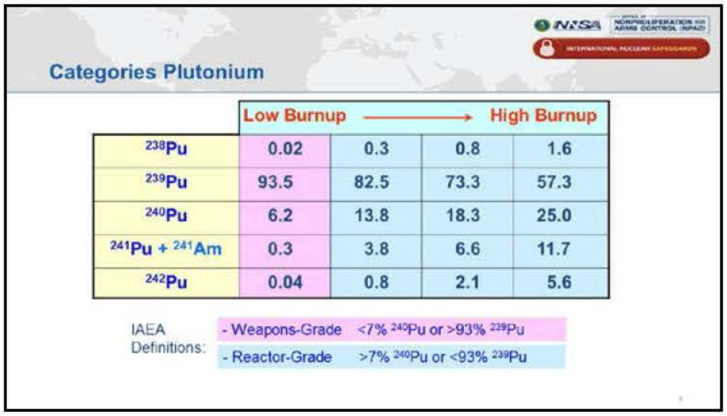


Plutonium does not exist naturally. Instead, it is created in reactors. Pu-239 is a decay product of Np-239, which is a decay product of U-239, which is created when a U-238 atom absorbs a neutron.
Pu-240 is created whenever Pu-239 absorbs a neutron. Thus, high burnup fuel will have more Pu-240 relative to Pu-239 than low burnup fuel (see next slide).

Instructor Notes:

All plutonium originates in nuclear reactors and is produced by capturing extra neutrons by ^{238}U to form ^{239}U, which then undergoes a series of decays to form ^{239}Pu. Plutonium does not exist in nature but if you take ^{238}U, which makes up the overwhelming fraction of natural uranium and bombard it with neutrons, some nuclei will absorb a neutron, transforming them into ^{239}U. This nucleus has too many neutrons to be stable, and decays by beta decay transforming one of the neutrons in the nucleus to a proton, which transmutes the ^{239}U into Neptunium, ^{239}Np. ^{239}Np, while more stable than ^{239}U, remains unstable and undergoes beta decay, resulting in ^{239}Pu which can be considered as stable. The isotopic composition of plutonium is affected by how long it stays in the reactor. Short exposure produce plutonium with very little ^{240}Pu and with very little plutonium being consumed by fission. Long exposure produce high ^{240}Pu concentrations, and a substantial portion of the plutonium produced is consumed by fission.

Additional Information for Students:



Categories Plutonium

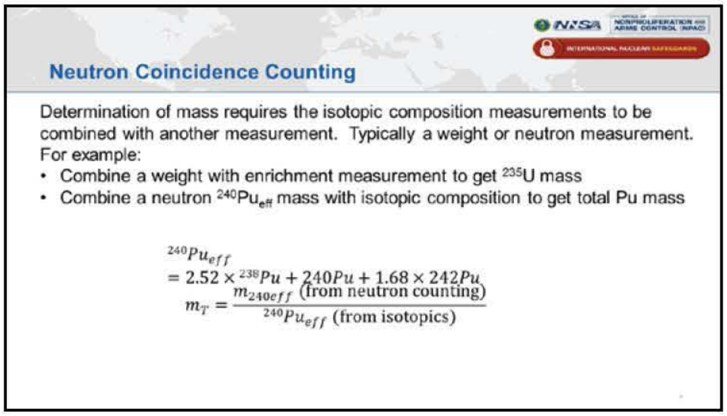
	Low Burnup	→	→	High Burnup
^{238}Pu	0.02	0.3	0.8	1.6
^{239}Pu	93.5	82.5	73.3	57.3
^{240}Pu	6.2	13.8	18.3	25.0
$^{241}Pu + ^{241}Am$	0.3	3.8	6.6	11.7
^{242}Pu	0.04	0.8	2.1	5.6

IAEA Definitions:
- Weapons-Grade <7% ^{240}Pu or >93% ^{239}Pu
- Reactor-Grade >7% ^{240}Pu or <93% ^{239}Pu

Weapons-grade plutonium (WGPu) can be extracted from low burnup fuel.

Instructor Notes:

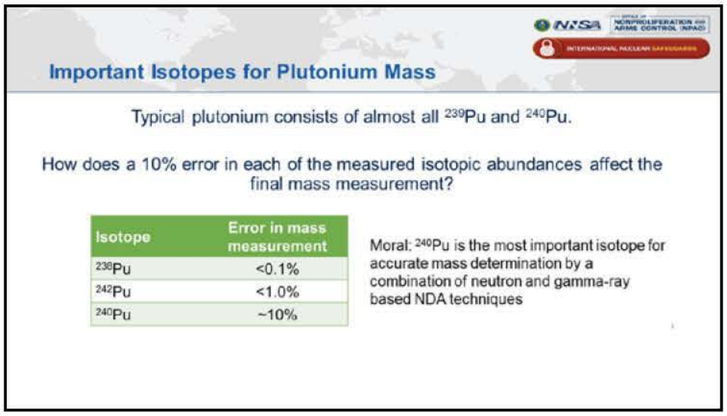
Additional Information for Students:



Instructor Notes:

Nuclear material assay aims to determine the mass of each isotope as an item, typically by combining gamma spectroscopy with neutron coincidence counting. All three of the even isotopes contribute to the response of a neutron-coincidence counter. The contribution from ^{240}Pu dominates for most plutonium bearing materials. Therefore the ^{240}Pu mass is defined by the equation, where ^{240}Pu eff is the mass of ^{240}Pu that would give the same coincidence response as that observed from the actual measured item.

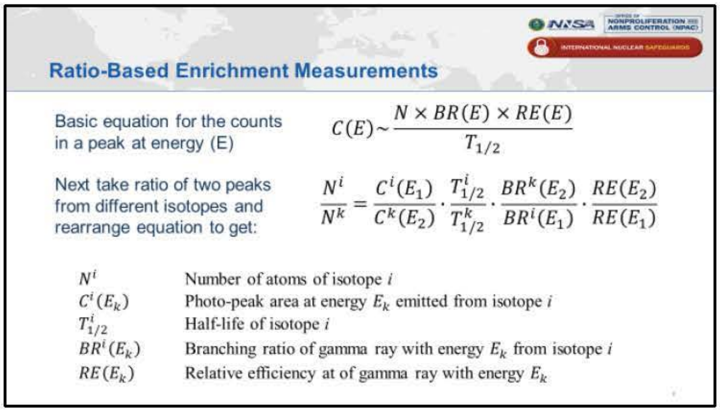
Additional Information for Students:



Instructor Notes:

Neutron coincidence counting measures spontaneous fission in Plutonium. Spontaneous fission yields many isotopes important for the fuel cycle, like ^{238}Pu, ^{240}Pu, ^{242}Pu. ^{240}Pu produces most of the spontaneous fission because is present in larger quantities, therefore, it is important to obtain accurate measurements of this isotopes. A 10% error in its measurement will cause a 10% error in the total Pu mass measurement. ^{238}Pu and ^{242}Pu have higher spontaneous fission rates but are present in smaller quantities; errors in their measurements have less effect on the total accuracy of Pu mass.

Additional Information for Students:



The number of counts in a peak is proportional to:

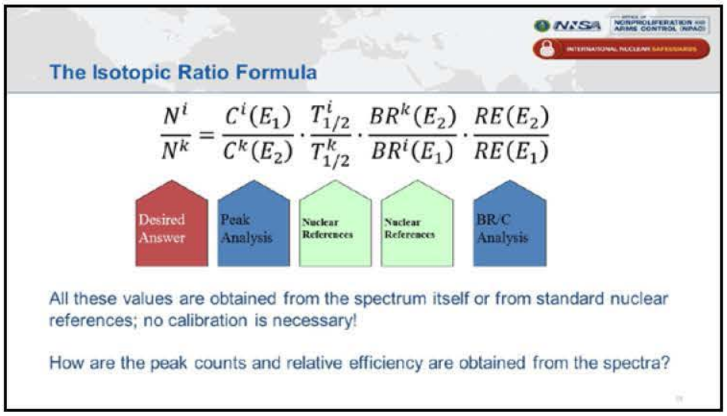
- The amount of material (N)
- The activity of the material, which is proportional to $1/T_{1/2}$
- The branching ratio (BR)
- The total efficiency, which is proportional to the relative efficiency RE

If you take the ratio of the number of counts in two peaks and rearrange, you can solve for the isotopic ratio.

Instructor Notes:

Standard expression for obtaining isotopic ratios from a gamma-ray spectrum. In this equation, the total efficiency has been rewritten in terms of the relative efficiency (RE), the geometric factors associated with the total efficiency cancel and relative efficiency includes the effects of sample self-absorption, attenuation in materials between the sample and the detector and detector efficiency. The need for only an efficiency ratio removes the problems associated with the geometric and sample reproducibility associated with absolute measurements and makes the method applicable to samples of arbitrary size, shape and composition.

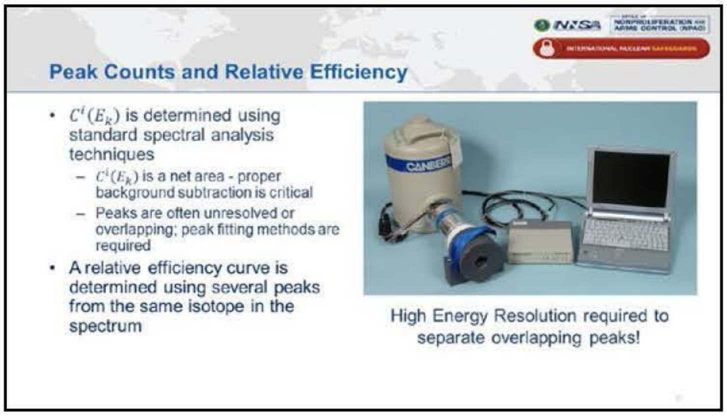
Additional Information for Students:



Notice that all of these values come from either the single measured spectrum or from nuclear reference data. Thus, we can find the isotopic ratio without any calibration measurements, without known geometries, and without prior detector characterization.

Instructor Notes:

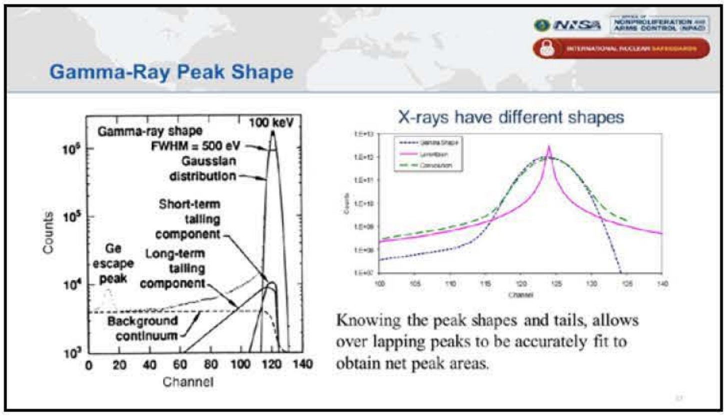
Additional Information for Students:



The first physical requirement for this technique is a high resolution detector. The peak information must be extracted very accurately, which will require an HPGe detector.

Instructor Notes:

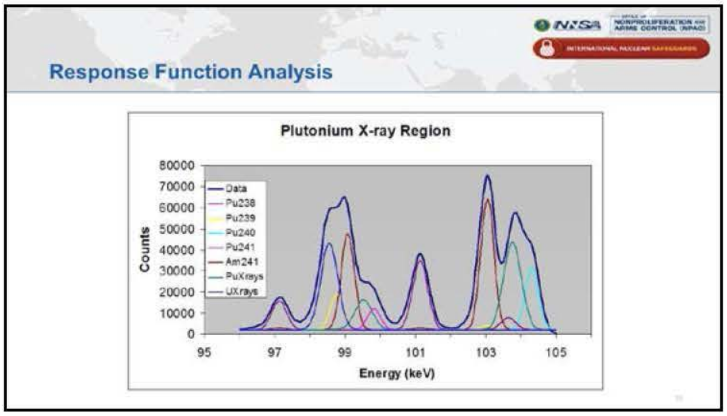
Additional Information for Students:



To accurately extract peak information, the real shape of the peaks must be known. Scattering (and poor electronic settings) can produce a low energy tailing on the peaks. Peaks from X-rays also have a fundamentally different shape, and while this is hard to manually observe even with HPGe detectors, it is a necessary consideration for peak fitting.

Instructor Notes:

Additional Information for Students:

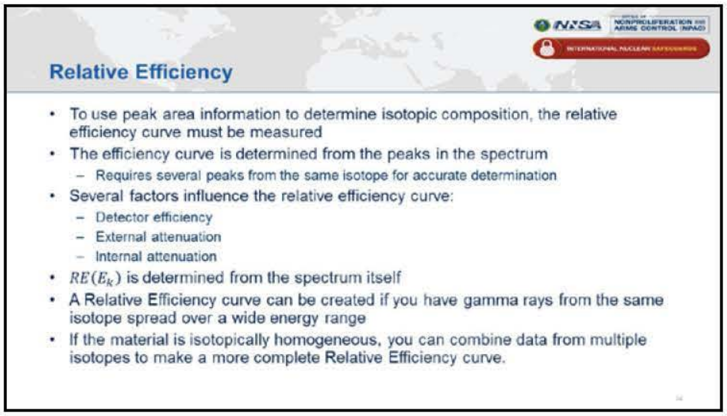


When analyzing unshielded plutonium/uranium, the x-ray region can be used for very accurate isotopic analysis. This region contains several useful gamma-rays, as well as several x-rays. Even with HPGe detectors, the peaks in this region overlap significantly and will require response function analysis to be useful.

Instructor Notes:

The Excel sheet that produced this simulated spectrum will be available to students in the lab. You can vary the grade of the plutonium to see the effect on this region.

Additional Information for Students:

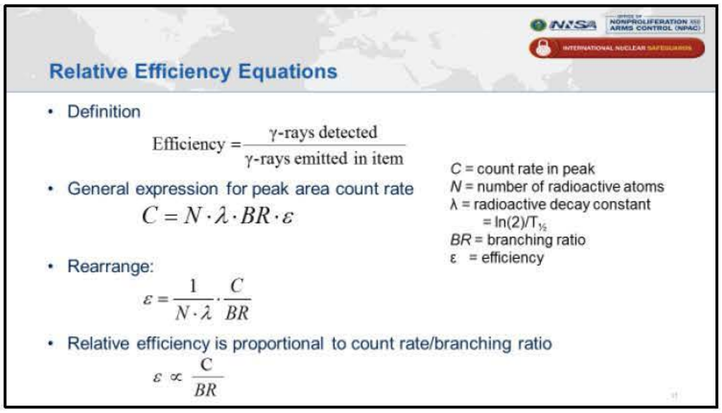


While knowing the total efficiency would be useful, it requires significantly more knowledge to calculate (detector efficiency, geometry, some material characteristics, and more). Instead, we can use the relative efficiency (RE) for this analysis, which removes many of these requirements.
The RE still includes the detector efficiency, attenuation effects, etc, but unlike the total efficiency it can be determined from the spectrum itself.

Instructor Notes:

Using relative efficiency can help simplify the analysis because it does not require ulterior measurements, it can be determined from the same spectrum being already analyzed. The relative efficiency is viewed as a function of energy. Almost any variable that perturbs the absorption or relative intensity of gamma rays emitted from the sample can affect the shape or energy dependence of the relative efficiency curve. The relative efficiency is proportional to the absolute efficiency and contains the detector efficiency, the external attenuation and the internal attenuation.

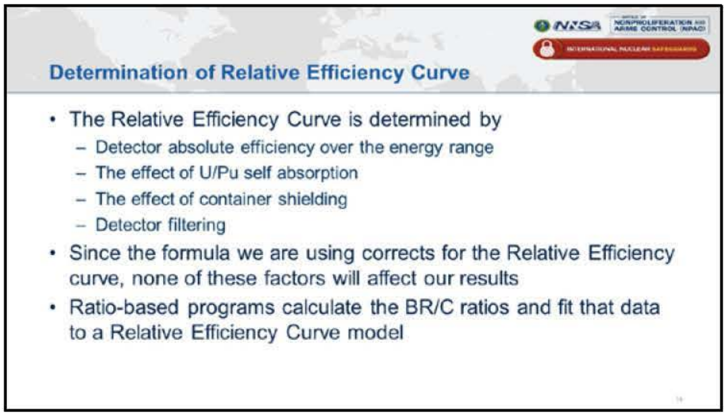
Additional Information for Students:



Thus, if we measure C for several peaks and divide each by their respective branching ratios, we obtain a measure of the relative efficiency.

Instructor Notes:

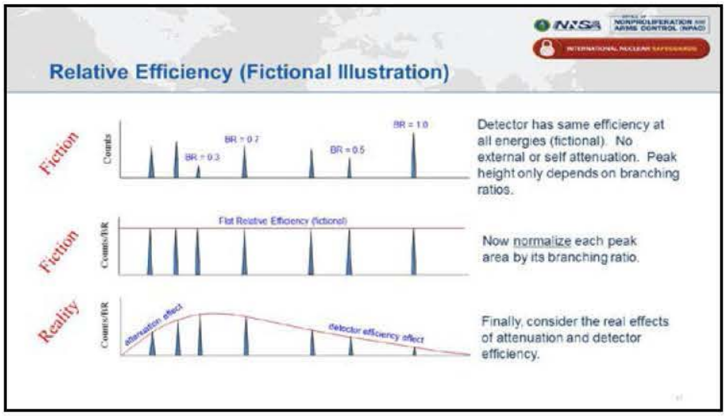
Additional Information for Students:



Instructor Notes:

The relative efficiency is determined from each measured spectrum by considering the energy variation of the quotient of BR/C for a series of gamma rays from a single isotope and then fitted to a relative efficiency curve model. The ratio takes into account the variations caused by the detector efficiency, the sample self-absorption and geometry, the container shielding and filters placed between the sample and the detector.

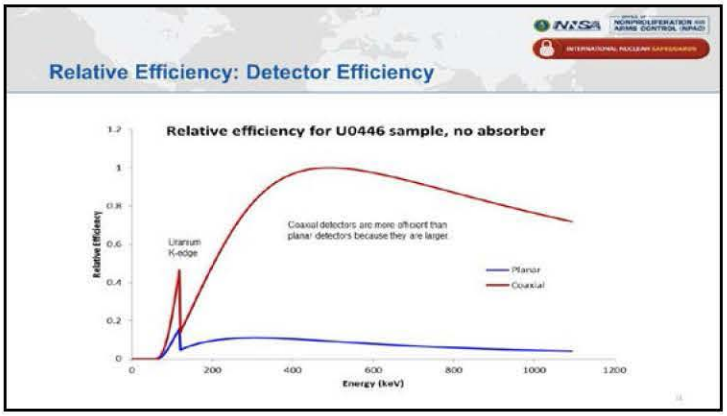
Additional Information for Students:



Consider this fictional example. In the top figure, a set of peaks is shown. If we divide the area of each peak by its branching ratio, we get the relative efficiency. If we did not have a uniformly efficient detector, we would likely observe an efficiency curve like in the last example.

Instructor Notes:

Additional Information for Students:

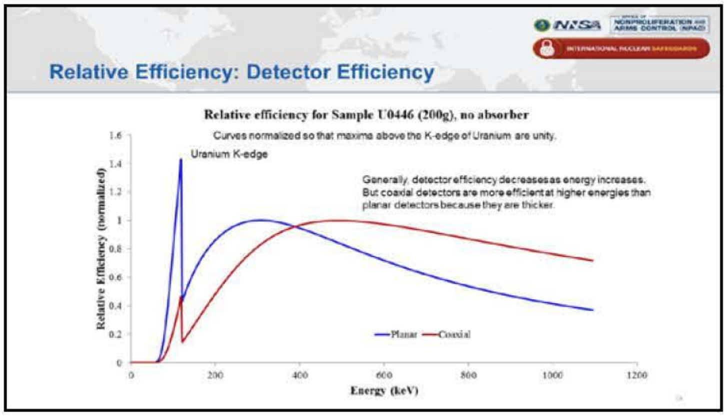


As mentioned, there are several components of the relative efficiency curve. First, it includes the detector's efficiency. Here, the coaxial detector is more efficient than the planar detector, particularly at higher energies.

Instructor Notes:

Question for the students: Why is the coaxial detector's efficiency so much higher?

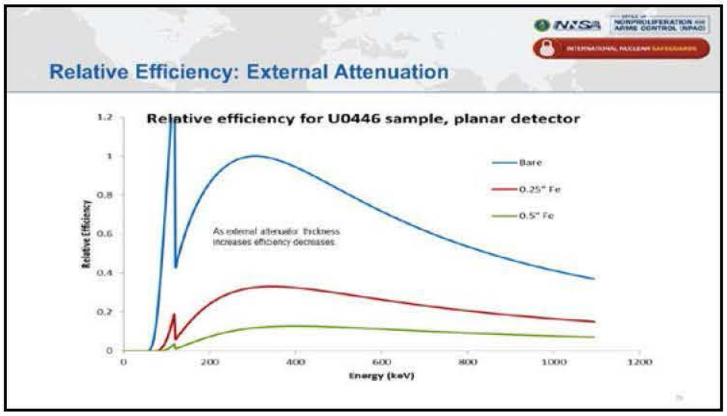
Additional Information for Students:



If we normalize each of the curves, we can better observe the relative difference between the two detectors.

Instructor Notes:

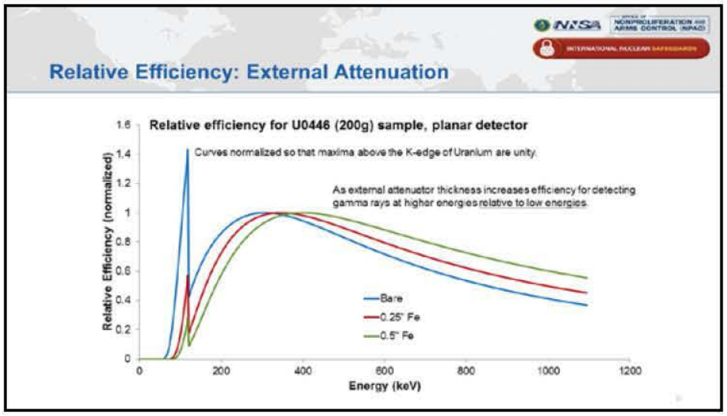
Additional Information for Students:



The next component of the relative efficiency curve is external attenuation. Shielding materials (lead bricks, steel containers, etc) have an energy-dependent shielding effect: low energy gamma-rays are much more likely to interact with them.

Instructor Notes:

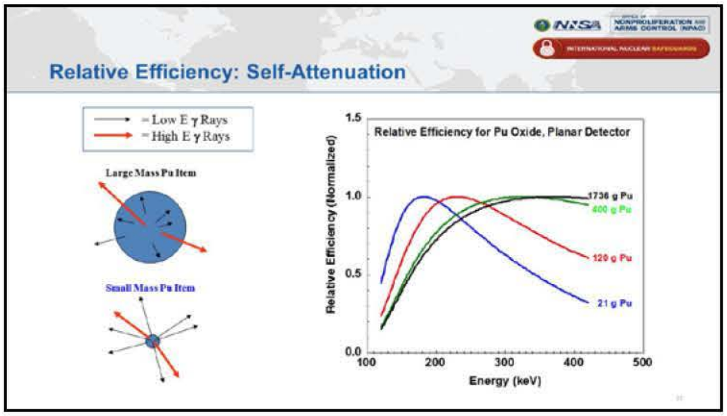
Additional Information for Students:



Comparing the normalized curves, it is clear that the effect of shielding is less at higher energies, and that shielded spectra may require using higher energy peaks for the analysis.

Instructor Notes:

Additional Information for Students:

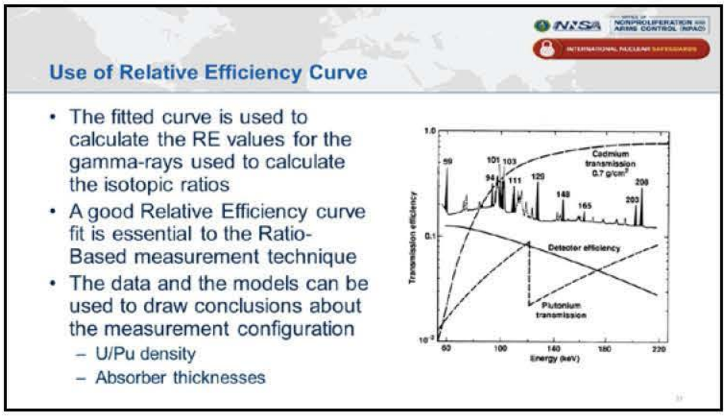


Finally, the source itself can attenuate any emitted gamma-rays. For very small items, this is negligible; for larger items, this effect becomes much more significant.

Instructor Notes:

The figure shows the overall effect seen as the sample gets larger, which usually also mean thicker. The curves are normalized to unity at their maximum value. The mean free path increases as energy increases, therefore, an isotopic measurement will see farther into the sample and hence more sample volume and mass at high energy than at low energy.

Additional Information for Students:

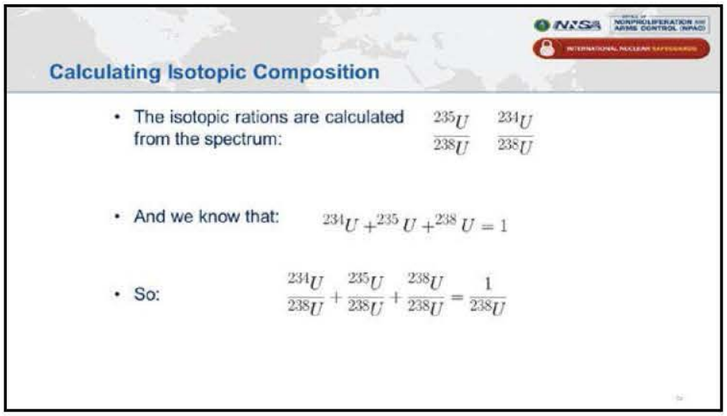


All of these effects combine to give an overall relative efficiency.

Instructor Notes:

Note that we didn't discuss geometric efficiency. Why?

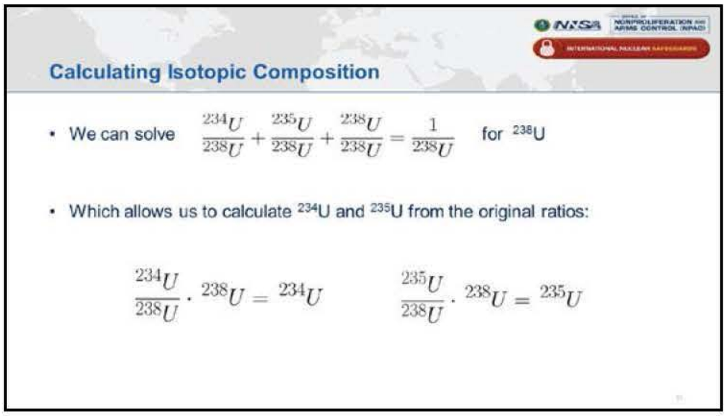
Additional Information for Students:



The result of this analysis is a set of isotopic ratios. We can convert this to isotopic fractions with a little arithmetic.

Instructor Notes:

Additional Information for Students:



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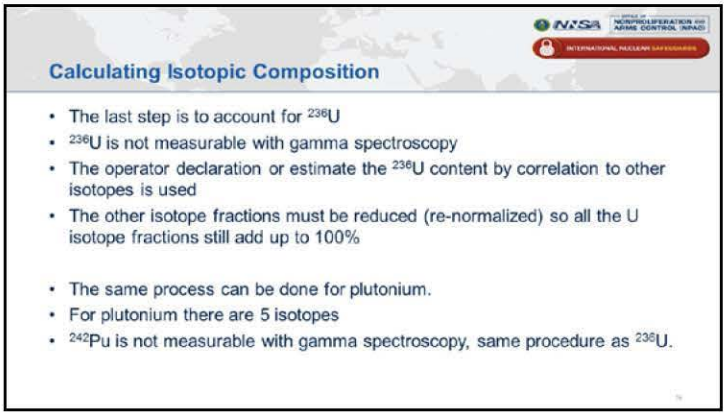

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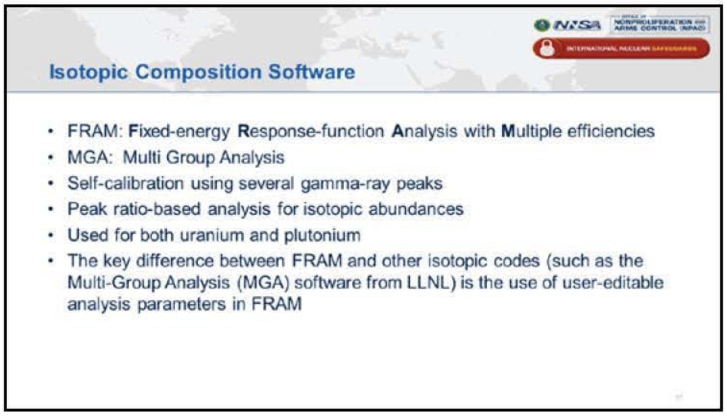
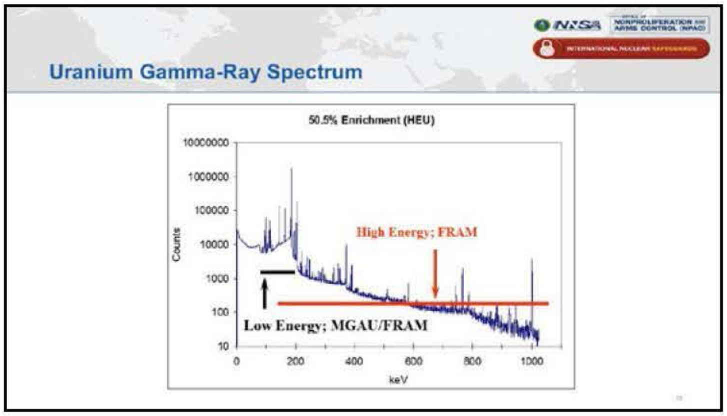


MGA and MGAU are similar software packages from LLNL; MGA analyzes plutonium, while MGAU analyzes uranium.

Instructor Notes:

There are multiple software able to measure isotopic composition.
FRAM: analyzes heterogeneous samples by creating two different efficiency curves, for the plutonium and the other material.
MGA is a widely used computer code for the analysis of high-resolution gamma ray spectra in order to extract the relative isotopic composition of plutonium for a diversity of items with minimal prior information.

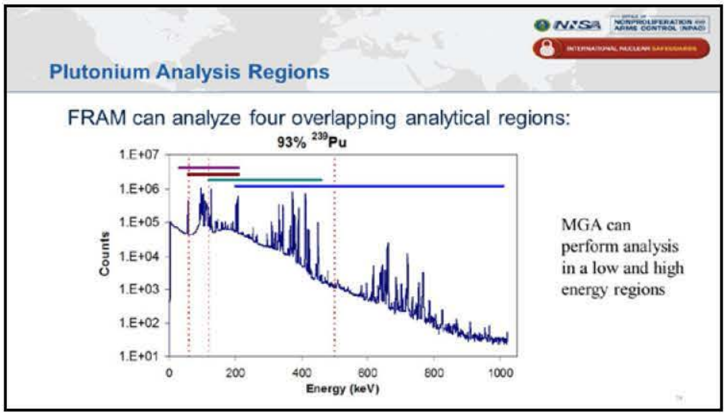
Additional Information for Students:



FRAM can use the low energy or the high energy region for the analysis. Older versions of MGAU are limited to low energy analysis.

Instructor Notes:

Additional Information for Students:



For plutonium, there are several different analysis regions that can be used. The detector and shielding configuration will dictate which region yields the best results.

Instructor Notes:

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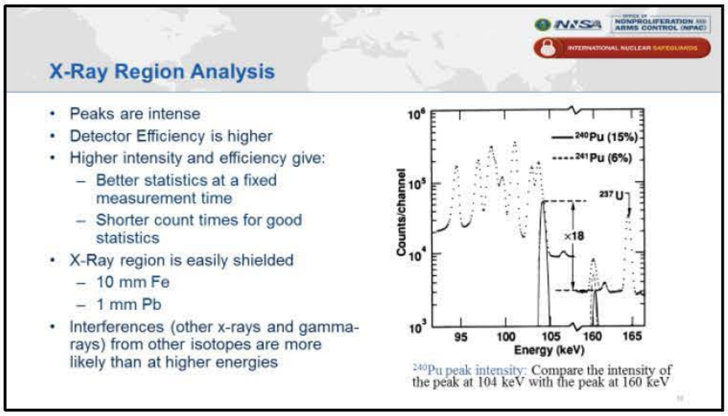
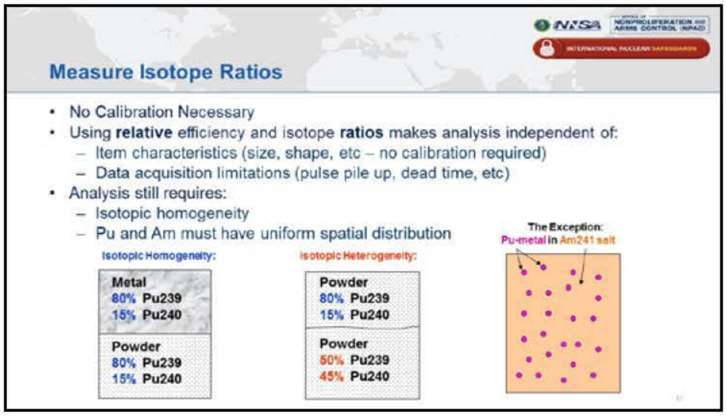


The x-ray region is (for unshielded items) typically the most intense region. It also has peaks for all of the isotopes of interest (aside from U-236 / Pu-242); this helps make the relative efficiency curve more reliable and simplifies the ratio analysis. However, this region can be easily shielded.

Instructor Notes:

Additional Information for Students:



For uranium, the infinite thickness method was a convenient way to measure the isotopics, but it

- Requires calibration
- Requires some knowledge of configuration
- Has geometry requirements
- Requires isotopic homogeneity
- Does not work for plutonium

Of these flaws, the isotopic ratio method only shares the homogeneity requirement.

Instructor Notes:

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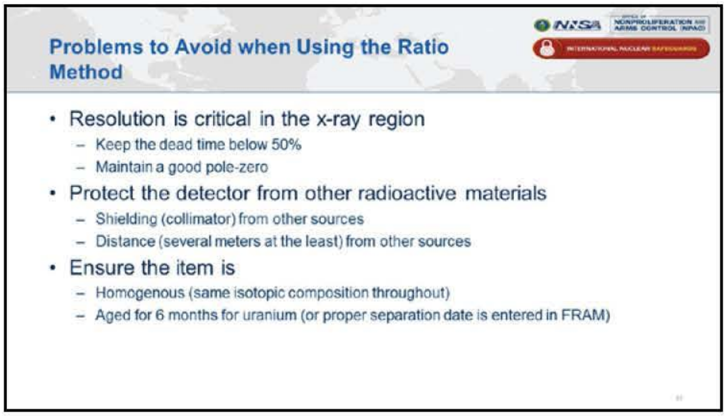

Fresh uranium can be analyzed with MGAU, but it will require a manual correction.

Instructor Notes:

Additional Information for Students:

Instructor Notes: